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Self-cleaning concrete doped with nano and micro-size zinc oxide particles

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ABSTRACT: *Nano- and micro scale zinc oxide (ZnO) particles were added to a concrete mix to study the effect on the self-cleaning ability of concrete given ZnO'photocatalytic abilities. Real sunlight was used to mimic environmental conditions, and two organic dyes (rhodamine B) and (methylene blue) were utilized as a model for organic pollutants in the air. ZnO particles were prepared in different concentrations (3, 6, 9, 12, 15%) in a proportional ratio to the cement. The results indicated that 6% microscale ZnO was the optimal amount to remove rhodamine B, with the removal efficiency reaching up to 94.5%, and 3% micro ZnO was optimal to remove methylene blue with removal reaching 87%. In contrast, 15% was the optimum concentration of nanoscale ZnO to remove both rhodamine B and methylene blue, with removal efficiency reaching up to 90% and 84.4%, respectfully. Scanning tunneling electron microscopy was used to see ZnO particle distribution on the surface of the prepared concrete.*
Keywords: *Zinc oxide, Self-cleaning concrete, Photocatalytic process, rhodamine B, methylene blue.*

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I. INTRODUCTION :-

Concrete is one of the most extensively used structural construction materials because of its affordability. It is the second most-consumed item in the world, after water [1]. Traditional concrete components are water, cement, fine and coarse aggregates (sand and gravel or crushed stones, respectively). When cement and water compound together, they make a paste, which binds all the aggregates together to make concrete [2]. Adding specific materials to the traditional mix of concrete can help improve its characteristics in order to help treat air pollution, one of the biggest environmental problems that have a considerable negative health impact [3].

Zinc oxide generally takes the form of a white powder and is almost insoluble in water. ZnO is similar to TiO2 and has global importance as an opacifier. It has the ability to absorb ultraviolet light (UV) and be utilized as a pigment in paints, paper coatings, sunscreen lotions, and plastics. Most importantly, ZnO is a photocatalyst that can remove organic compounds from polluted air and water and be used for microbial disinfection [4,5]. Because ZnO has a similar bandgap energy to TiO2, their photocatalytic absorption is also expected to be comparable[6]. ZnO typically generates an electron-hole pair under solar irradiation, which leads to the formation of free radicals. ZnO has many beneficial characteristics, including being biosafe, environmentally safe, chemically stable in basic and acidic mediums, and relatively low cost. All these factors make it an advantageous filler in cementitious materials with the power to improve the antimicrobial nature and self-cleaning efficiency of concrete under sunlight [9].

Furthermore, using ZnO in mortar can help improve the characteristics of mortar. By adding ZnO as a partial replacement of cement 1, 3, and 5% by weight of cement. The compressive and tensile strength were tested at 7 and 28 days. The results showed that the mechanical characteristics of samples containing 3% and 5% ZnO were better than traditional mortar. Scanning electron microscopy (SEM) indicated that ZnO micro- and nanoparticles totally filled the pores and sped up the hydration process of cement particles, enhancing the durability and mechanical strength of the ZnO-cement mortar specimens [10].

Researchers recorded that the compressive strength of ZnO-doped concrete was around 18% greater than the traditional concrete. Side-by-side durability and density of concrete also improved [11]. Other researchers studied the effect of ZnO on the compressive strength and setting time of self-compacting concrete paste and found that adding ZnO improved both characteristics [12].

Researchers have also studied the use of ZnO as a photocatalytic agent by mixing cement with different concentrations (5, 10, 15%) of ZnO and testing the photocatalytic removal of organic pollutants represented by rhodamine 6G under UV irradiation[13]. Additionally, they studied the ability of ZnO-cement mixes to remove different kinds of bacteria. The cement-ZnO composite showed effective antifungal and antibacterial behavior under solar light and even in the dark[14].

In this study, ZnO nano- and micro-size particles were added to a concert mixture and explored as photocatalytic agents to remove organic pollutants, represented in a specific model with rhodamine B (red) and methylene blue (blue). Concrete mixed with certain amounts of both nano- and micro-size particles exhibited an exceptional ability to remove around 90% of the organic materials within 2-3 days of sunlight exposure.

II. EXPERIMENTAL DETAILS

2.1 Materials

2.1.1 Cement

Type I Portland cement was purchased from Ash Grove Cement Company and used in all mixtures to avoid differences between samples. The properties of the cement, as obtained from the company, are shown in Table 1.

Table 1. Portland cement properties, per the company.

Component	Percent by weight
SiO2	20.08%
Al2O3	4.65%
Fe2O3	4.11%
CaO	63.63%
MgO	0.94%
SO3	3.19%
Na2O	0.16%
K2O	0.54%
Limestone	2.7%

2.1.2 Coarse aggregate

The coarse aggregate was purchased from Webco Mining, Inc. The coarse aggregate complied with ASTM C-136.15grading requirements; absorption capacity: 1.2%, specific gravity:2.57. Table 2 lists the gradation of the coarse aggregate.

Table 2. Coarse aggregate gradation obtained from the company Webco Mining, Inc.

Sieve size	% Passing as tested
1.5 inch (38 mm)	100
¾ inch (19.05 mm)	95.1
3/8 inch (9.5 mm)	28.55
# 4 (4.75 mm)	5.2
# 8 (2.36 mm)	0.4
# 16 (1.18 mm)	0.3

2.1.3 Fine aggregate

The fine aggregate (sand) (specific gravity: 2.62, absorption capacity: 0.48%) was purchased from Jeffery Sand Co. It complied with ASTM C-3315, as Table 3 shows. All the information obtained from the company.

Table 3. Fine aggregate gradation as obtained from the company Jeffery Sand CO.

Sieve size	% Passing as tested
3/8 inch (9.5mm)	100
# 4 (4.75 mm)	97
# 8 (2.36 mm)	86
# 16 (1.18 mm)	80
# 30 (600 micro meter)	45
# 50 (300 micro meter)	13
# 100 (150 micro meter)	0.5

2.1.4 Water

All concrete mixes were prepared with clean, fresh tap water without impurities.

2.1.5 ZnO particles

Both nanoscale (100 nm) and micro-scale (5 micrometer)ZnO with purity up to 99.9% were obtained from Sigma Aldrich.

2.1.6 Dyes

0.01 millimoles of rhodamine B dye and 0.01 millimoles of methylene blue dye were obtained to model organic pollutants. Both red and blue dyes were utilized to provide visual contrast.

2.2 Methods -

In this research, a proportional ratio of nano- and micro scale ZnO to cement (3, 6, 9, 12 and 15%) was used in each concentration. Three samples were prepared side by side while making the control mix, which contained the traditional concrete components (water, fine aggregate, coarse aggregates and cement) in order to compare the ordinary mix of concrete with the ZnO-infused concrete mixes. After mixing the dry ingredients together completely, water was added and the mixtures put in molds for 24 hours. After the concrete samples were prepared, similar procedures as previously reported were followed to test the samples' self-cleaning abilities [15]. Briefly, 0.1 ml of rhodamine B and 0.1 ml of methylene blue dye were applied on the top of the concrete samples, and the samples were left to let the dye set. Next, the samples were placed in sunlight to reflect real usage conditions; all the samples were placed together to prevent unwanted variations in results. The self-cleaning ability of the control and doped concrete was measured by calculating the color intensity degradation after 1, 2, 3 and 4 days. ImageJ® software was utilized to study the color removal efficiency and determine the top-performing ZnO size and dose. Table 5 details the contents of the ZnO concrete mixes and control mix.

Table 5. Components of the different concrete mixes used in this study.

Material (lb/yd3)	Control	3%	6%	9%	12%	15%
Cement	730	708	686	664	642	620
Fine Aggregate	1994	1994	1994	1994	1994	1994
Coarse Aggregate	983	983	983	983	983	983
Water	400	400	400	400	400	400
ZnO	-----	22	44	66	88	110

III. RESULTS

Both the nano- and micro-scale ZnO degraded the dyes on top of the concrete; however, the micro-ZnO performed better than the nano-ZnO. The optimal dose to remove rhodamine B was 6% micro-ZnO, removing 94.5% of the color, and the optimum dose to remove methylene blue was 3% micro-ZnO, which removed 87% of the color. The optimal amount of nano-ZnO was 15%, which removed 89.7% of the RhB color and 84.62% of MB dye. Figure 1-A shows rhodamine B dye degradation for 6% micro-ZnO, and Figure 1-B shows methylene blue dye degradation for 3% micro-ZnO. Figures 1-C and figure 1-D present rhodamine B and methylene blue degradation, respectively, by 15% nano-ZnO 1,2,3, and 4 days after exposure to real sunlight. Results suggest that nano-and micro scale ZnO-infused concrete can remove organic pollutants on its surface. Figures 2 and 3 present the rhodamine B and methylene blue removal efficiency, respectively, for all ratios of micro-ZnO. Figures 4 and 5 show nano-ZnO's removal efficiency for rhodamine B and Methylene blue respectfully.

While 6% micro-ZnO removed 94.5% of the rhodamine B, 3% removed 93% of the rhodamine B. Thus, we recommend using 3% micro-ZnO as the optimum dose because there was not a significant difference between 6% and 3% in removing Rhodamine B, and using less ZnO is more cost effective. Figure 6-A shows the SEM image of this optimum dose of ZnO, and Figure 6-B shows the scanning tunneling electron microscope image and the purple dots represent the distribution of the optimal does of micro-ZnO particles on the surface of the concrete sample . In order to be used in practical applications, the mix simply needs to be deposited as the top layer of concrete, ~10% of the thickness of the overall composite, as only the surface is engaged during photocatalysis.

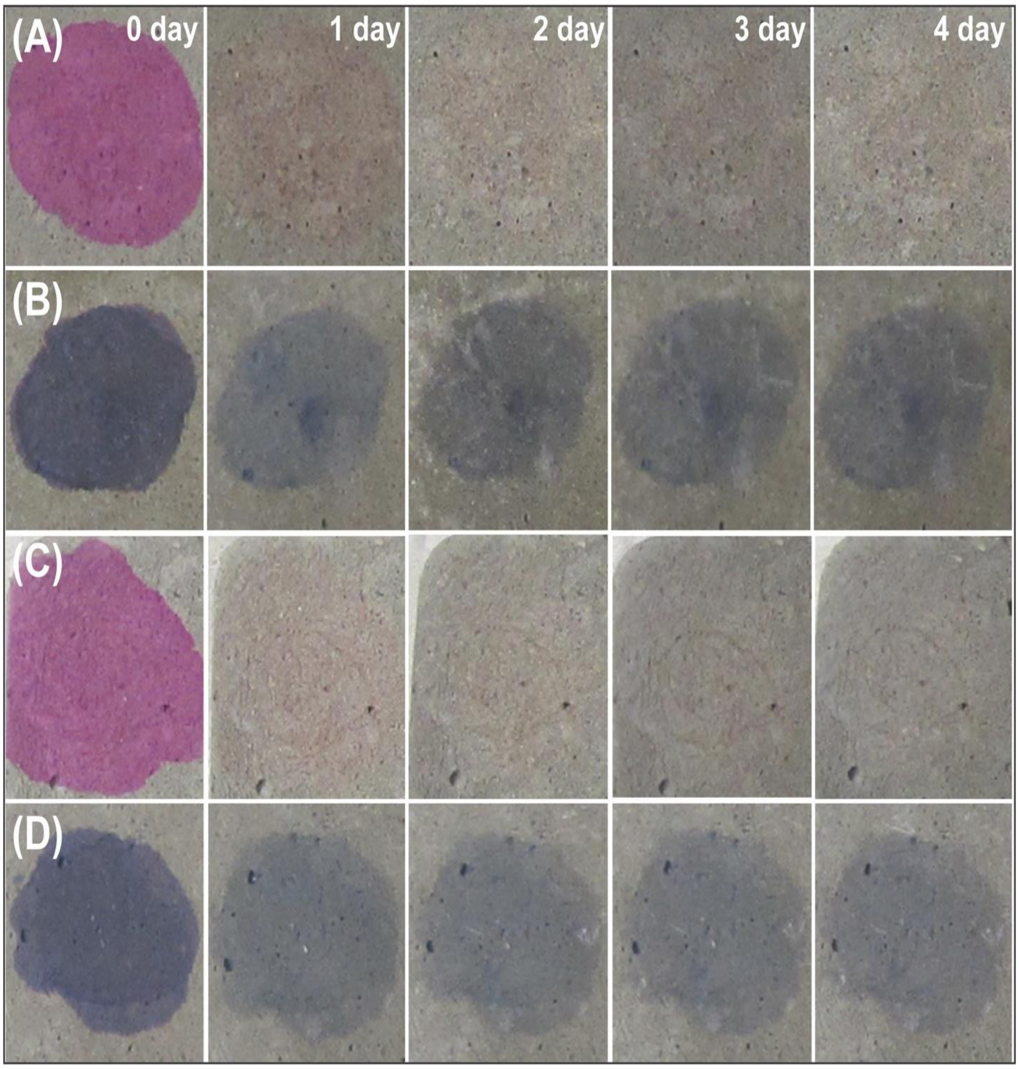

Figure 1. A)Rhodamine B degradation by 6% micro-ZnO at 0, 1, 2, 3, and 4 days.B)Methylene blue degradation by 3% micro-ZnO at 0, 1, 2, 3, and 4 days. C)Rhodamine B degradation by 15% nano-ZnO at 0, 1, 2, 3 and 4 days. D)Methylene blue degradation by 15% nano-ZnO at 0, 1, 2, 3, and 4 days.

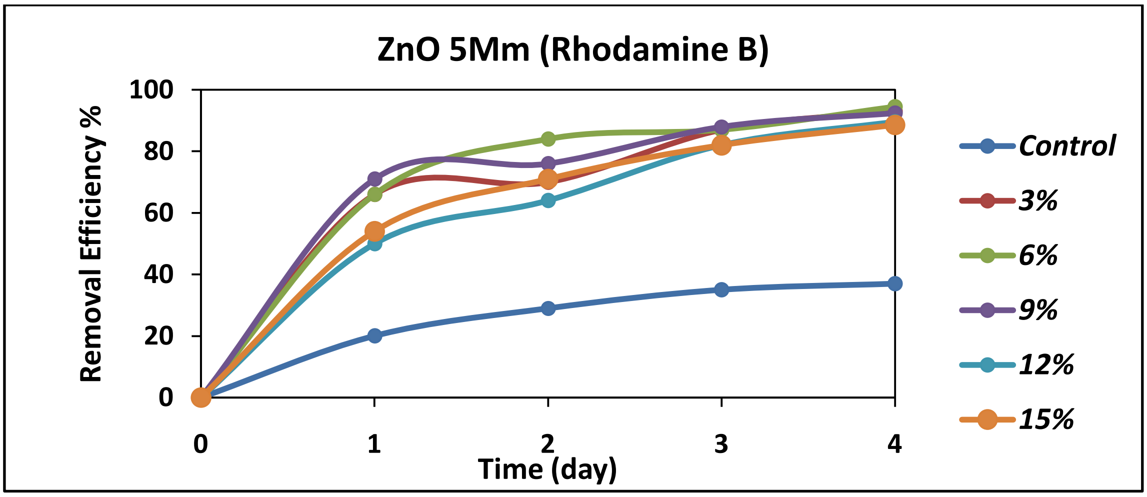

Figure (2) Rhodamine B removal efficiency for 5 Micro ZnO at (3%, 6%, 9%, 12%, 15%) and control mix of concrete with time (day)

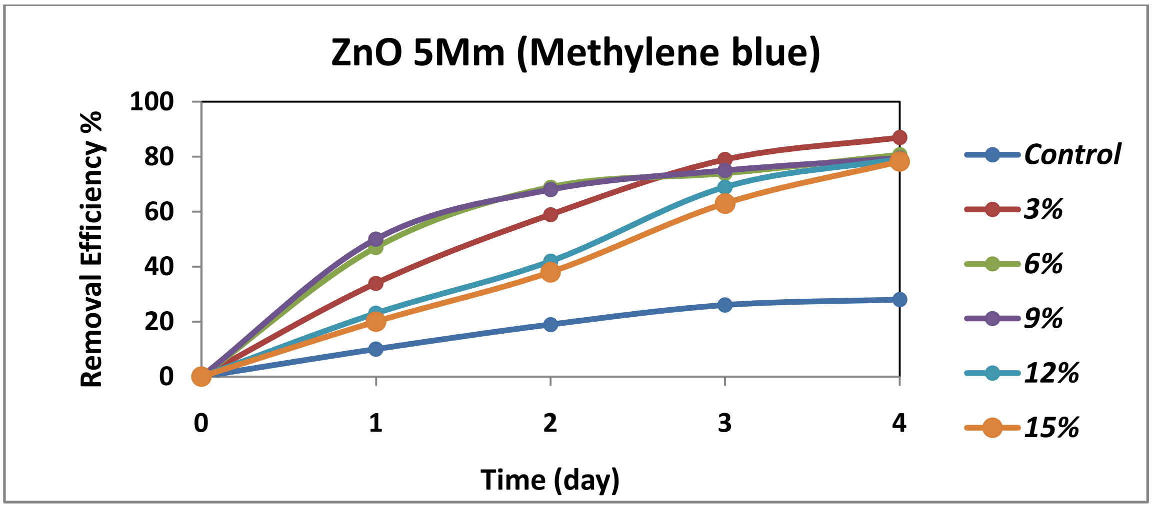

Fig (3) Methylene blue removal efficiency for 5 Micro ZnO at (3%, 6%, 9%, 12%, 15%) and control mix of concrete with time (day)

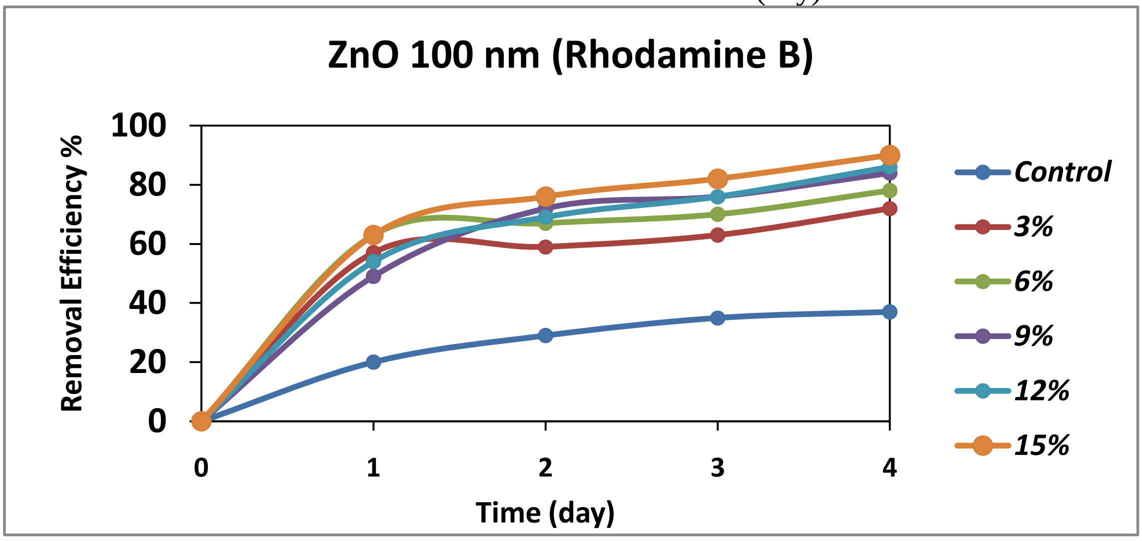

Fig (4) Rhodamine B removal efficiency for 100 nano ZnO at (3%, 6%, 9%, 12%, 15%) and control mix of concrete with time (day)

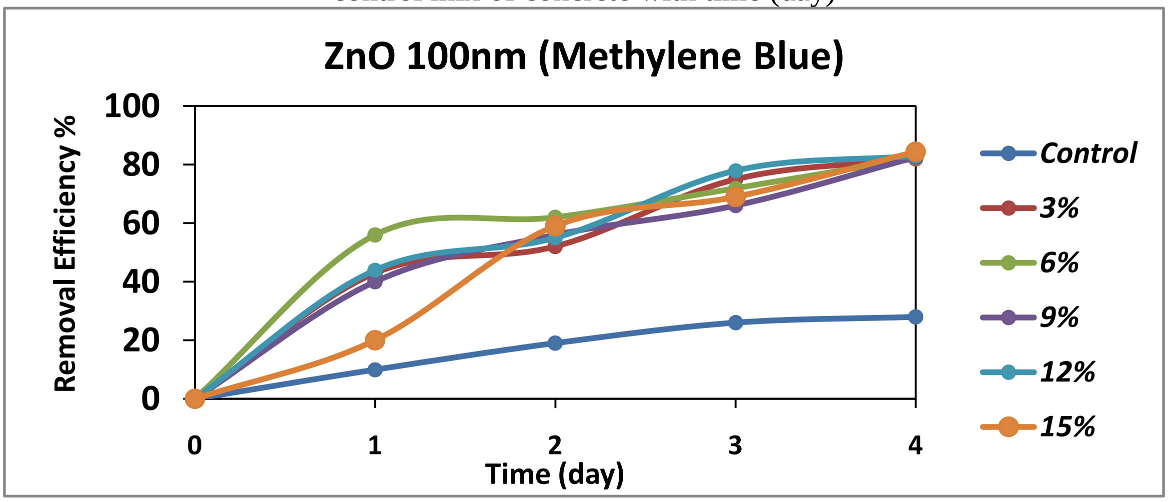

Fig (5) Methylene blue removal efficiency for 100 nano ZnO at (3%, 6%, 9%, 12%, 15%) and control mix of concrete with time (day)

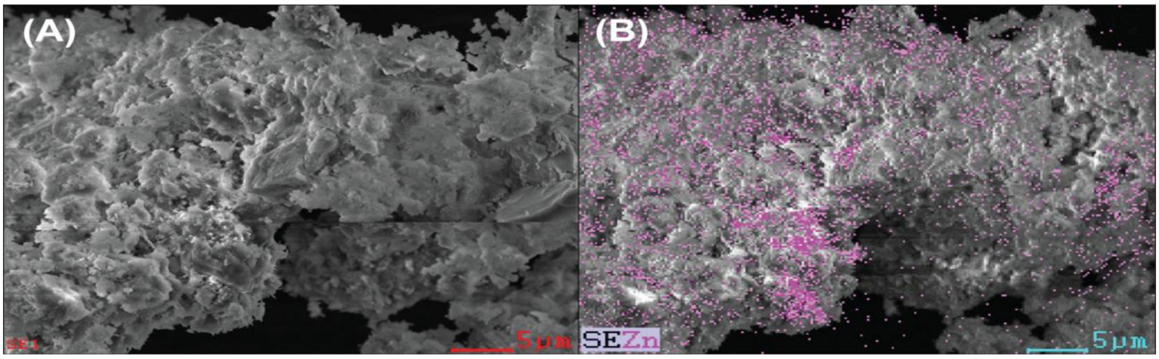

Figure 6.A) Scanning electron microscope image of concrete with 3% infusion of micro-size ZnO particles;B)scanning tunneling electron microscope image of concrete with 3% infusion of micro-size ZnO particles; purple dots represent the distribution of ZnO particles inside the concrete structure.

IV. CONCLUSION

The self-cleaning ability of micro- and nano scale ZnO particles were studied in detail. Results showed that adding a certain amount of ZnO led to the removal of organic materials on the surface of the concrete by more than 90% when exposed to sunlight for several hours. The two organic pollutant models, rhodamine B and methylene blue, were successfully removed within several hours due to the photocatalytic activity of the ZnO particles. Based on our results, we recommend using 3% micro-ZnO as the optimum dose to remove pollutants from the surface of the concrete.

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